

Review

Overview of Molecular Structure Design of Polymeric Functional Binders for Lithium-Ion Battery Cathodes

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Abstract: Polymer binders for the cathodes of lithium-ion batteries play a pivotal role in enhancing the battery energy density, cycle life and interfacial stability. Conventional polyvinylidene fluoride (PVDF) binders have difficulty meeting the multiple requirements of high specific energy electrodes due to their inherent drawbacks including electrical insulation, mechanical rigidity and interfacial inertness. In recent years, function-oriented binders have emerged as a research hotspot, which are endowed with properties such as electrical conductivity, mechanical adaptability, self-healing capability and high voltage tolerance via rational molecular structure design. This review systematically reviews the molecular design strategies of polymer-based cathode binders, including the introduction of polar groups (e.g., hydroxyl and carboxyl groups) to strengthen hydrogen bonding and electrostatic interactions, the construction of conjugated polymers or ion-conductive networks to improve electrical conductivity, and the utilization of dynamic chemical bonds to achieve self-healing function. In addition, high voltage-tolerant binders can significantly suppress capacity fading of high-voltage cathodes via chelating transition metal ions, optimizing electrode dispersibility and forming a stable interfacial layer. By integrating molecular dynamics simulations with experimental characterizations, the correlation between the molecular structure of binders and the electrochemical performance of electrodes is elucidated, which provides a theoretical framework for the design of next-generation high-performance binders.

Keywords: cathode; polymer binder; lithium-ion batteries; structural design

1. Introduction

With the ever-growing demand for high-efficiency energy storage devices, lithium-ion batteries (LIBs) have attracted increasing attention owing to their high energy density, excellent rate performance, long cycle life, and environmental benignity [1,2]. As core devices for clean energy storage and conversion, they have been widely applied in electric vehicles, smart grids, and portable electronic devices. Driven by the continuous pursuit of higher energy density, emerging cathode materials such as high-nickel ternary (NCM/NCA) materials, lithium-rich manganese-based (LRMO) materials, and sulfur cathodes have become research hotspots [3]. However, these materials encounter severe challenges during the charge-discharge process: interfacial side reactions of high-nickel cathodes [4,5], polysulfide shuttling of sulfur cathodes [6–8], and other issues, all of which lead to electrode structural damage and rapid capacity decay. Against this backdrop, polyvinylidene fluoride (PVDF) [9,10], a conventional commercial binder, despite its good chemical and electrochemical stability [11,12], only binds electrodes via intermolecular interactions and thus fails to provide strong adhesion [13–15]. Furthermore, its insulating nature results in high impedance, making it difficult to meet the multiple requirements of high specific



energy electrodes for the stability of conductive networks, adaptability to volume expansion, and interfacial protection functions.

Cathode materials play a key role in lithium-ion battery performance. As the component accounting for the largest proportion of weight and cost in lithium-ion batteries, they occupy more than one-third of the total battery cost and remain one of the main bottlenecks for achieving high battery performance. Although binders only account for 2–5% of the electrode mass [16], they play a central role as an “invisible skeleton” in regulating electrode microstructure: their functions are not limited to bonding active materials and current collectors, but also directly affect the continuity of the internal conductive network, the uniformity of stress distribution, and ion transport kinetics in the electrode. The limitations of traditional PVDF-based binders are particularly prominent in high-performance battery systems—for instance, their insulating property compels the addition of excessive conductive agents (e.g., carbon black, with a proportion up to 10–20%) in the electrode [17], sacrificing the active material loading; their rigid segments [18] cannot adapt to the volume expansion of specific active materials, leading to electrode pulverization and abrupt capacity decline; their chemical inertness makes it difficult to inhibit interfacial side reactions between the cathode and electrolyte under high voltage, accelerating battery failure. In recent years, researchers have broken through traditional cognition and proposed the design concept of “function-oriented binders”: multiple properties are endowed via molecular engineering, transforming them from passive bonding to active function regulation. For example, conductive binders can replace part of conductive agents to improve electrode energy density; dynamic cross-linked networks can repair microcracks during cycling through bond rearrangement to extend electrode lifespan; binders containing polar functional groups can anchor transition metal ions and block the chain reaction of electrolyte decomposition. This transformation from “inert binders” to “multifunctional interfacial engineering materials” not only provides a new idea for the development of high specific energy batteries, but also promotes the innovation of battery material design, possessing profound scientific significance and industrialization value.

Based on the aforementioned context, we focus on the multifunctional molecular design strategies of polymer-based cathode binders and systematically review the breakthrough progress in recent years. Following the main line of “functional demand–molecular design–performance enhancement”, this review first analyzes implementation pathways the basic functions such as conductivity enhancement, mechanical adaptability and interfacial stabilization. For example, the synergistic effect between polymer binders and conductive nano-networks [19] is utilized to construct dual electron/ion transport channels, or cross-linked networks are employed to disperse local stress. It then extends to emerging directions such as self-healing and high voltage tolerance, clarifying the action mechanisms of dynamic bonding and intelligent responsive groups. By integrating molecular dynamics simulations, *in-situ* characterizations and battery performance data, this review aims to establish a quantitative correlation between the molecular structure of binders and the macroscopic performance of electrodes, thereby providing a theoretical framework for the rational design of next-generation lithium-ion battery binders.

2. Molecular Design Strategy of Multifunctional Cathode Binder

2.1. Enhanced Bonding Properties

As a crucial component that tightly connects active materials and conductive agents to form a robust conductive network, and generates strong interactions with current collectors to ensure stable adhesion of electrode components to the collectors [20], the most essential function of binders is their bonding capability, which is closely related to the cycle life and discharge capacity of batteries. To this end, numerous studies have been conducted to enhance the adhesive capacity of binders. It is a consensus in the scientific community that introducing polar groups to strengthen interactions is an effective approach to improve bonding performance. The incorporation of polar groups can enhance the integrity of the entire electrode structure, thereby reinforcing the interactions between binders, current collectors, and active materials [21].

2.1.1. Forming Hydrogen Bonds

When hydrogen atoms are attracted to other electronegative atoms (e.g., oxygen and nitrogen atoms), intramolecular or intermolecular hydrogen bonds can be formed [22]. Such bonds are usually introduced by incorporating corresponding functional groups, such as hydroxyl (–OH) and carboxyl (–COOH) groups. For instance, carboxymethyl cellulose (CMC) [23], polyacrylic acid (PAA) [24], alginates, and other materials possess abundant –OH and/or carboxylate (–COOR) groups, which can form strong hydrogen bonds with active materials.

For example, Jiang et al. [25] designed a fluorine-free organic–inorganic hybrid cross-linked siloxane-based binder (SNH). The strong hydrogen bonding interactions formed between the abundant silanol groups (Si–OH) in the binder and the electrode components significantly enhanced the adhesion strength between the Ni-rich cathode

particles and the current collector, and promoted the formation of uniform electron/ion transport pathways even at high mass loading. Jiao et al. [26] synthesized a cyanoethyl-functionalized chitosan binder (CSAN). The cyano groups ($-\text{C}\equiv\text{N}$), amino groups ($-\text{NH}_2$), and hydroxyl groups ($-\text{OH}$) in its molecular structure interact with each other to form a dynamic three-dimensional hydrogen-bond network, which greatly enhanced the adhesion capability and constructed an efficient lithium-ion transport channel. Duan et al. [27] designed a composite binder (H-P) by integrating hydrogenated nitrile butadiene rubber (HNBR) and poly(acrylic acid) (PAA), as shown in Figure 1. The abundant $-\text{COOH}$ groups in PAA formed strong hydrogen bonds with the $-\text{OH}$ groups on the electrode surface, which significantly enhanced the adhesion. As a result, the peel strength of the H-P electrode reached 23.23 N m^{-1} , much higher than that of PVDF (0.322 N m^{-1}). After cycling, the electrode remained intact without cracks. Meanwhile, the strong Li^+ coordination ability of the $-\text{COOH}$ groups promoted the formation of a LiF-rich cathode electrolyte interphase (CEI), thus accelerating the reaction kinetics. Compared with the PVDF-based counterpart (40.3% capacity retention after 500 cycles at 1 C), the LiFePO_4 (LFP) cathode based on H-P exhibited significantly better capacity retention (86.3% after 500 cycles at 1 C).

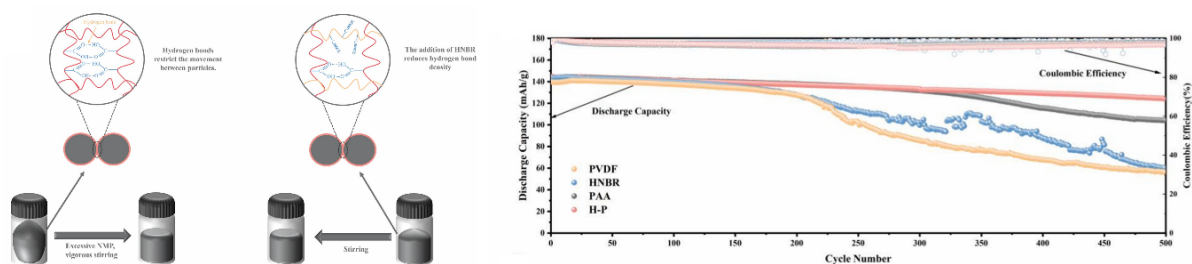


Figure 1. Schematic illustration of binder distribution during slurry mixing and Long-term cycling performance at 1 C (500 cycles) [27]. (Reprinted with permission from Ref.[27]. Copyright 2026, Elsevier Inc.).

Polyimide (PI) is a type of polymer with outstanding thermal stability and excellent mechanical properties, which has been widely used in gas separation [28,29], memory materials [30,31], hydrogen storage [32], and other fields. In addition, it also serves as a novel functional binder with excellent thermal properties, high adhesive strength, and diverse structural designability [33,34]. Using PI as a cathode binder (e.g., copolyimide (P84)) can not only maintain similar electrochemical performance to PVDF binders, but also improve the safety of lithium-ion batteries (LIBs) [29,35,36].

Given the excellent bonding performance of polyimide, numerous studies have been conducted on the application of PI in battery cathodes, especially those focusing on introducing polar groups to form hydrogen bonds for enhancing bonding capacity. For example, Wang et al. [37] synthesized two types of polyimide (PI) binders containing different polar functional groups (PI-OH and PI-COOH). By introducing strongly polar functional groups to form hydrogen bonds, the interaction between electrode components was enhanced. When applied in NCM cathode materials, the binders enabled excellent long-term cycling performance at a high voltage of 4.7 V. The initial discharge specific capacity was 205 mAh/g at a current density of 40 mA/g, and the capacity retention was 73% after 100 cycles at 4.7 V, which was much better than the 34% capacity retention of PVDF.

In contrast, Liang et al. [38] synthesized a cross-linked polyimide (PI) binder (PI-APE-AZ). The strong hydrogen bonds were mainly formed between the polar imide groups ($\text{C}=\text{O}$, $-\text{NH}-$) in the molecular chains and the oxygen atoms or hydroxyl groups on the surface of the cathode materials, which significantly enhanced the adhesion strength of the electrode. The peel strength reached 0.531 N/mm, more than 10 times higher than that of PVDF (0.051 N/mm), which relies only on weak van der Waals forces.

2.1.2. Forming Electrostatic Interactions

In lithium-ion battery cathodes, electrostatic interactions dominate the binding between binders and active materials through Coulombic attraction between charged groups (e.g., $-\text{SO}_3^-$ or $-\text{N}^+\text{R}_3$) and ions on the surface of active materials (e.g., Li^+ or transition metal ions), thereby enhancing the structural stability and interfacial adhesion of electrodes. In addition, electrostatic interactions further improve the cycling performance and safety of batteries by regulating slurry dispersibility (preventing agglomeration of active materials and conductive agents) and inhibiting interfacial side reactions (such as transition metal dissolution and electrolyte decomposition).

Jeong et al. [39] investigated the application of poly(vinylidene fluoride-chlorotrifluoroethylene) (PVdF-CTFE) as a binder, with a focus on the effect of its polar groups on lithium-ion adsorption and transport. As illustrated in Figure 2, the polarized electron environment formed by the electronegative F atoms in PVdF-CTFE

can facilitate electrostatic interactions with Li^+ , enabling uniform and tight contact among active materials, solid electrolytes, and conductive agents. Compared with the nonpolar BR binder, PVdF-CTFE can construct more efficient Li^+ diffusion pathways.

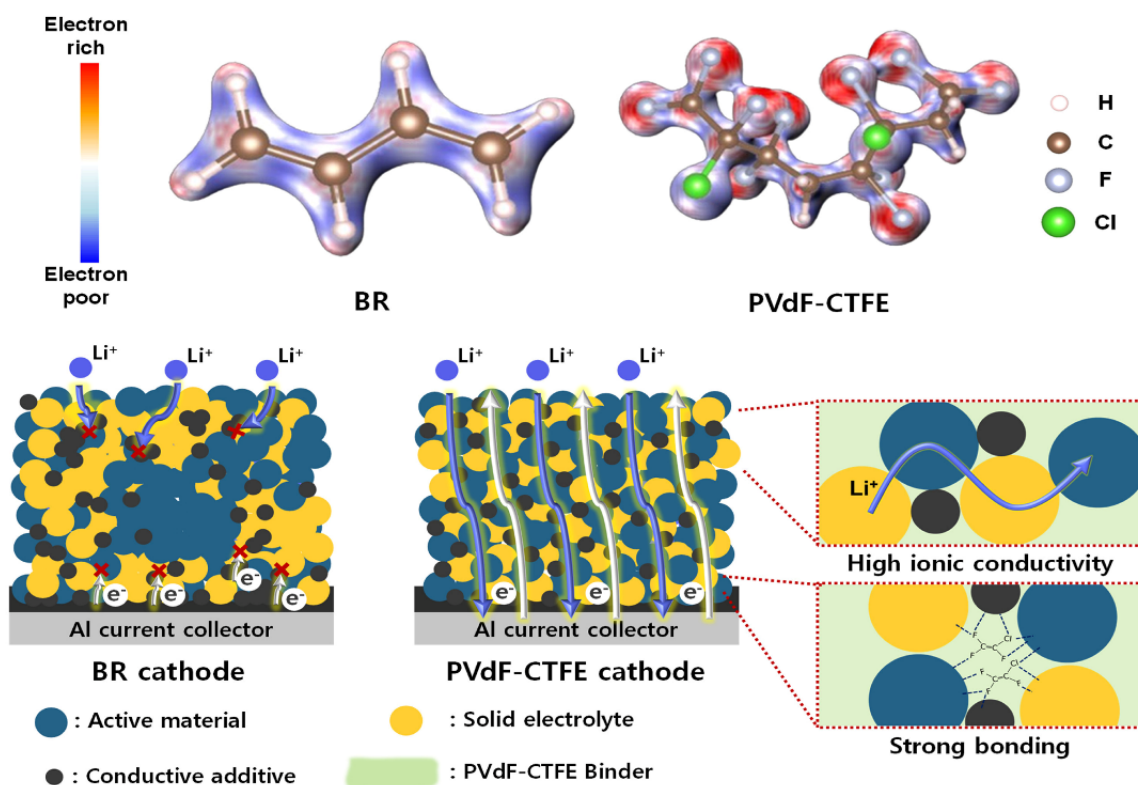


Figure 2. Schematic illustration of charge density mapping about each binder and the binder effect on adhesion and Li^+ diffusion properties in the slurry based composite cathode [39]. (Reprinted with permission from Ref. [39]. Copyright 2024, Elsevier Inc.).

Among them, zwitterionic polymers (ZIPs) [40,41], composed of oppositely charged cationic and anionic moieties attached to the polymer backbone or side chains, have been used as electrolytes in lithium-ion batteries [42,43]. These ZIPs containing charged and polar moieties are ion-conductive, facilitating Li^+ transport [44,45]. Furthermore, the charged and polar moieties in ZIPs are expected to enhance their adhesion to active materials in lithium-ion battery cathodes, thereby maintaining the structural stability of electrodes during long-term operation and extending battery lifespan. For example, Yang et al. [46] synthesized a series of zwitterionic polymer (ZIPs) binders via the copolymerization of poly(ethylene glycol) methyl ether methacrylate (PEGMEA) and sulfobetaine methacrylate (SBMA). As shown in Figure 3, after mixing ZIP-9 with LFP, the binding energy peaks of the sulfonate groups shifted from 167.0/168.1 eV to 167.2/168.3 eV, confirming the existence of strong electrostatic interactions between the zwitterionic polymer and the LFP surface. The 180° peel test in Figure 3b further quantified the adhesive strength: ZIP-9 reached 0.61 N cm^{-1} , approximately 15 times that of PVDF. Moreover, the adhesive strength first increased and then decreased with increasing SBMA content. These two lines of evidence collectively elucidate the molecular mechanism by which cationic and anionic groups in the zwitterionic structure synergistically enhance interfacial adhesion.

Ionic bonds can be formed by two oppositely charged ions located in different polymers or ionic linkers, and their binding capacity is usually stronger than that of hydrogen bonds. The binding capacity is mainly determined by the properties of charged structures, such as diameter, valence, ion concentration, polymer structure, and terminal functional groups [17], with interaction strength higher than that of general electrostatic interactions. For instance, Wang et al. [47] designed a layer-by-layer stacked structure between negatively charged Nafion and positively charged polyvinylpyrrolidone (PVP), which can generate strong electrostatic interactions. They found that the peel force of this structure was much higher than that of PVDF.

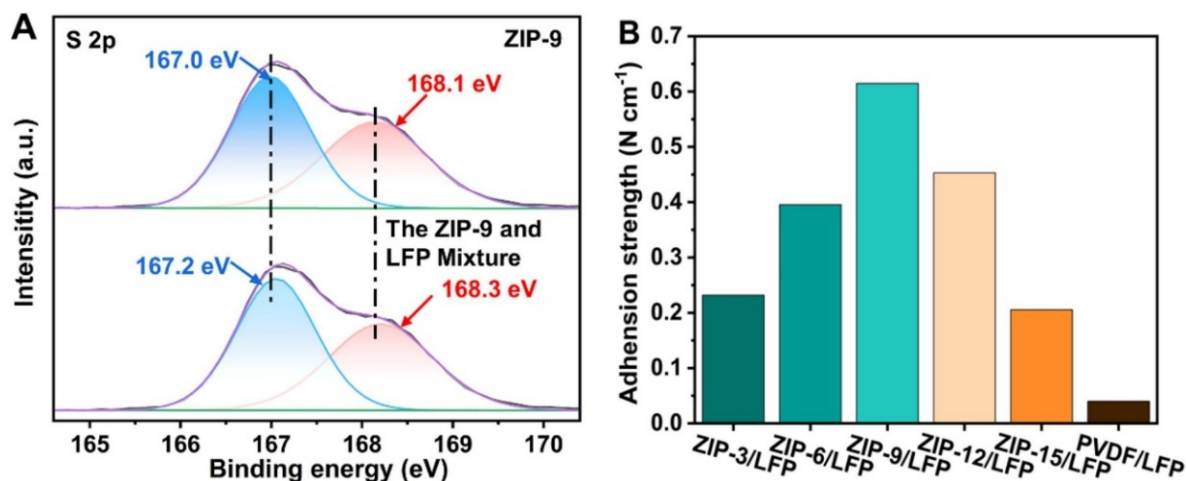


Figure 3. S 2p XPS spectra of the ZIP-9 and LFP mixture and pure ZIP-9 (A), adhesion strength of the ZIPs/LFP and PVDF/LFP cathodes (B) [46]. (Reprinted with permission from Ref. [46]. Copyright 2025, Elsevier Inc.).

2.1.3. Other Forces

In addition to the aforementioned hydrogen bonds and ionic bonds, there are numerous other molecular structure design strategies to enhance the adhesive capacity of binders. For example, polyvinylidene fluoride (PVDF), as a commercially available binder, has been widely used; however, the interaction between PVDF and current collectors is van der Waals force, which is weak and results in low bonding strength. To address this issue, researchers have referenced the regulation of polymer crystallinity via heat treatment [48,49] to improve bonding strength. The Kim group [50] heated PVDF at 220 °C to control its crystallinity and found that the heated electrode exhibited higher discharge specific capacity, and the capacity retention was 83.7% after 100 cycles at 0.5 C. Correspondingly, the enhanced bonding strength was verified by scanning electron microscopy (SEM) and electrochemical impedance spectroscopy (EIS).

Another strategy is the formation of cross-linked structures. By constructing three-dimensional networks, this approach fundamentally solves the problems of traditional linear binders (e.g., PVDF) in cathodes, such as insufficient bonding strength, poor electrolyte swelling resistance, and low interfacial stability. Wang et al. [51] developed a composite binder featuring a dynamically cross-linked network, which established multiple highly efficient lithium-ion (Li⁺) transport pathways through the cross-linked structure. Zhou et al. [52] used wheat gluten (WG) as a binder, where a small amount of water promoted hydration to trigger intermolecular and intramolecular disulfide bonds, leading to self-cross-linking of the material and thus enhancing its bonding efficiency. In contrast, Luo et al. [53] developed a biomass lignin-based 3D cross-linked polymer binder (PNAL), which was prepared by cross-linking alkaline lignin (AL) and poly(acrylonitrile-co-acrylic acid) (PNA). Owing to the formation of a 3D cross-linked network structure, this structure can tightly wrap electrode particles, fix the interface, significantly enhance the adhesion to the current collector, and improve the overall bonding performance. Thus, as shown in Figure 4, the PNAL electrode exhibits the highest bonding strength, and its swelling resistance to the electrolyte is also improved. In terms of battery performance, the PNAL-based LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂ (NCM811) lithium metal battery (LMB) achieved a high capacity retention of 75.7% after 300 cycles at 3 C under 4.6 V.

The formation of coordination bonds serves as a reference strategy for the molecular design of most binders for NCM cathodes. As mentioned earlier, NCM active materials undergo phase transitions and dissolution-migration of transition metal ions during operation, resulting in reduced battery cycle life and specific capacity [54]. Therefore, scientists often introduce polar groups into polymer molecules when designing binders for NCM electrodes to form coordination interactions with NCM particles, alleviate structural damage, and trap dissolved transition metal ions to maintain capacity. For instance, the Jeong group [55] introduced carboxyl groups into the polynorbornene backbone, which form hydrogen bonds with –OH on the surface of NCM active materials to enhance interactions. As shown in Figure 5, the bonding capability is higher than that of PVDF. Furthermore, it can be seen that simply increasing polar groups leads to uneven distribution due to excessively high surface energy, thus the principle of “bonding-dispersion” balance needs to be considered. Besides carboxyl groups, polar groups such as hydroxyl, urethane, and nitrile groups are also popular choices. Jin et al. [56] simultaneously introduced carboxyl, hydroxyl, amide groups, and other polar groups into the polymer, significantly enhancing the bonding capacity. Moreover, spandex can also be used as a highly elastic binder in NCM cathodes; its alternating soft-hard segment structure enables uniform coating on the cathode surface to protect the structure. For example, Lee et al. [57] used spandex as

a binder, utilizing polar groups such as ureido and amino groups in it to coordinate with dissolved metal particles and reduce capacity loss. Xu et al. [58] employed polyacrylonitrile (PAN) as a binder, leveraging the carbon-nitrogen triple bonds in PAN to form coordination bonds with transition metal ions [59], thereby immobilizing them.

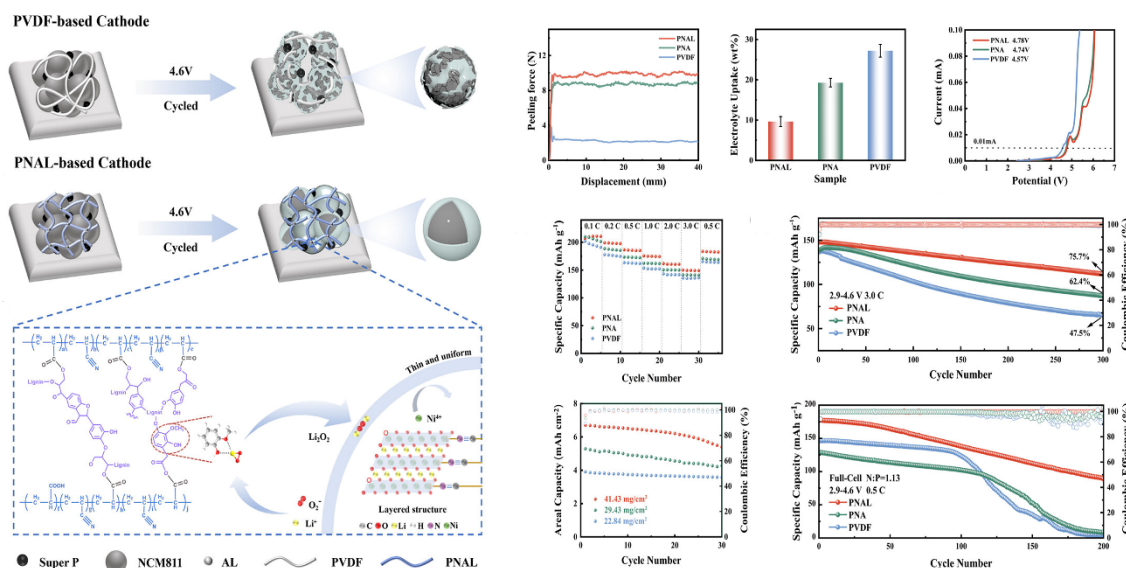


Figure 4. Illustration diagram of PNAL-assisted interface engineering and mechanisms; mechanical and electrochemical performance of PNAL binder [53]. (Reprinted with permission from Ref. [53]. Copyright 2026, Wiley-VCH.).

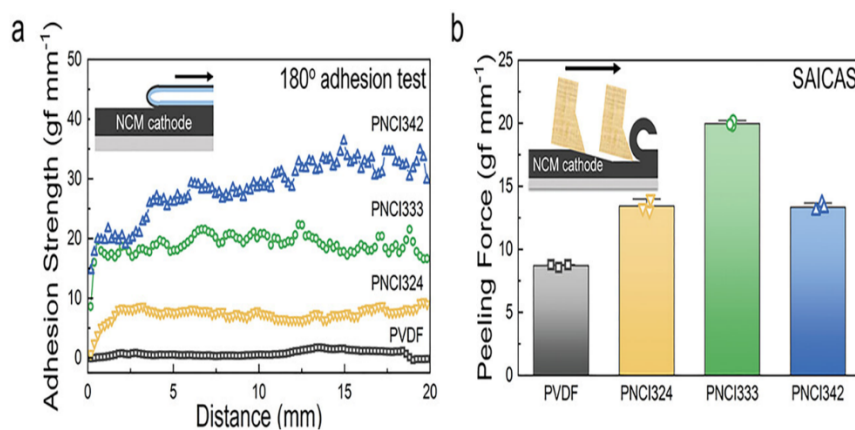


Figure 5. Adhesion and dispersion properties of NCM cathodes prepared with PVDF and PNCI# binders (NCM: Super P: binder = 90:5:5 wt%). (a) 180° adhesion test profiles of the NCM cathodes with PVDF or PNCI# binder. (b) Peeling force values measured by SAICAS [55]. (Reprinted with permission from Ref. [55]. Copyright 2023, Wiley-VCH.).

Moreover, polyimide (PI) can also play a role in ternary positive electrodes, and its coordination effect can improve battery performance. For example, Pham et al. [60] introduced fluorine-containing groups into the PI main chain; the -COOH groups in the polymer can coordinate with transition metal ions, thereby enhancing the bonding strength. Meanwhile, the fluorine element improves the electrochemical stability and oxidation resistance of the polymer, effectively suppressing the decomposition of the electrolyte under high-voltage conditions. Chen et al. [61] designed a novel polyimide binder with a high dielectric constant and sulfonyl groups for NCM (nickel-cobalt-manganese) positive electrodes. The strongly polar amide groups (-CONH-) and sulfonyl groups (-SO₂-) in the binder molecule contain oxygen atoms with lone pairs of electrons, which can form coordinate bonds with the empty orbitals of transition metal ions (Ni, Co, Mn) on the surface of the NCM811 particles. This coordination interaction not only significantly improves the bonding strength of the electrode (the peel strength increased from 0.151 N·mm⁻¹ for PVDF to 0.571 N·mm⁻¹), but also enhances the structural stability of the electrode. Electrochemical tests showed that the NCM811 cathode using this binder exhibited a capacity retention of 78.6% after 100 cycles at 4.7 V and 0.2 C, which was much higher than that of the PVDF-based electrode (only 52.8%).

In addition, at a 5 C rate, the discharge capacity of the PI-based electrode reached $164.4 \text{ mAh}\cdot\text{g}^{-1}$, while that of the PVDF-based electrode was only $118.9 \text{ mAh}\cdot\text{g}^{-1}$, indicating that the PI binder effectively promotes ion transport and improves battery rate performance.

2.2. Enhanced Ionic/Electronic Conductivity

The working principle of rechargeable lithium-ion batteries is based on electrochemical reactions, and their performance (especially rate capability) is mainly related to ion and electron transport [62,63]. In electrodes, binders and conductive agents (e.g., conductive carbon black and carbon nanotubes) establish channels for electron and ion transport to facilitate redox reactions in electrodes, where binders bond the entire conductive network to maintain its integrity and continuity. However, this inevitably increases the loading of inactive components, leading to a significant decrease in battery energy density [64]. Therefore, in addition to bonding capability, preparing binders with excellent electron and ion transport properties is an effective approach to reduce the proportion of inactive substances and improve battery energy density. Generally, conductive binders can be divided into two categories: electronic conductive binders and ionic conductive binders, and there are various chemical methods to finely regulate the molecular structure by introducing different atoms and functional groups.

2.2.1. Ionically Conductive Polymer Binder

Traditional binders (e.g., PVDF) are ionic insulators. Ionic conductive binders require the formation of lithium-ion transport channels through material design to facilitate Li^+ diffusion between cathode active particles and reduce interfacial impedance. To this end, there are three molecular design strategies for such binders: first, introducing polar groups—incorporating polar groups containing oxygen, nitrogen, and other elements (e.g., polyethylene oxide (PEO), polyacrylonitrile (PAN)) into polymer chains to promote ionic conduction via coordination interactions with Li^+ . Second, constructing ionic channels by using block copolymers to form a continuous ionic conductive phase. Third, doping lithium salts such as LiTFSI into binders to enhance ionic conductivity (e.g., PVDF-HFP/LiTFSI composite system).

For example, Tsao et al. [65] synthesized two types of polyimides with sulfonic acid groups and ether groups, respectively: SPI (sulfonated polyimide), an ionic single-ion conductor containing sulfonate groups (SO_3^-), and EPI (ether-based polyimide), a coordination-type conductor that coordinates with Li^+ through oxygen atoms. Tests showed that SPI can form a wide diffusion double electric layer, reduce interface polarization, and improve high-rate performance, with a lithium-ion diffusion coefficient (D) of $7.23 \times 10^{-10} \text{ cm}^2/\text{s}$; in contrast, EPI strongly coordinates and fixes Li^+ through oxygen atoms, leading to multiple diffusion layers and high polarization, with a Li^+ diffusion coefficient of $5.49 \times 10^{-11} \text{ cm}^2/\text{s}$, which is lower than that of SPI but still much higher than that of PVDF ($6.10 \times 10^{-13} \text{ cm}^2/\text{s}$). Hong et al. [66] copolymerized tetrafluoroethylene with a perfluoromonomer containing $-\text{SO}_3^- \text{Li}^+$ side chains to prepare a single-ion conductive ionomer binder, which was applied to all-solid-state dry cathodes. It was found that the polymer had an ionic conductivity of $1.6 \times 10^{-5} \text{ S}\cdot\text{cm}^{-1}$ at 25°C , and the ionic conductivity of the composite cathode was $4.1 \times 10^{-3} \text{ S}\cdot\text{cm}^{-1}$, which was superior to that of the PTFE-based cathode ($2.9 \times 10^{-3} \text{ S}\cdot\text{cm}^{-1}$). The discharge specific capacity reached $180.7 \text{ mAh}\cdot\text{g}^{-1}$ at 0.1 C, and the capacity retention rate was 90% after 300 cycles at 0.5 C. Das et al. [67] introduced oligoether segments into the side chains of PProDOT-Hx₂ polymer to promote Li^+ transport, and found that when the oligoether side chain content was optimal, the ionic conductivity of the polymer reached $4 \times 10^{-7} \text{ S}\cdot\text{cm}^{-1}$, which was four times higher than the previously reported ionic conductivity of PProDOT-Hx₂ ($1 \times 10^{-7} \text{ S}\cdot\text{cm}^{-1}$). Similarly, Rodrigo et al. [68] synthesized a π -conjugated polymer with oligoether side chains and applied it to high-load ($\approx 14 \text{ mg}\cdot\text{cm}^{-2}$) and low-electrolyte ($E/C \approx 3 \text{ g}\cdot\text{Ah}^{-1}$) battery systems. It was found that the polymer with ion-conducting oligoether side chains had the highest ionic conductivity ($\approx 10^{-6} \text{ S}\cdot\text{cm}^{-1}$) and moderate porosity, achieving a high energy density of $323 \text{ Wh}\cdot\text{kg}^{-1}$ at C/2 rate, which was much higher than that of PVDF-based batteries ($250 \text{ Wh}\cdot\text{kg}^{-1}$).

Considering the lithium-ion conductivity of polyethylene oxide (PEO), numerous studies have focused on synthesizing polymers containing PEO segments for use as electrode binders. For instance, Kuo et al. [69] synthesized a fluorinated copolymer by copolymerizing 2-(perfluorohexyl) ethyl methacrylate (PFHEMA) and poly(ethylene glycol) methyl ether methacrylate (PEGMA), and used this PEO-based copolymer as a binder for LiFePO_4 (LFP) cathodes, and the molecular dynamics simulations revealed that PEO segments can reduce the association between Li^+ and PF_6^- and increase the number of free Li^+ ions. However, they can also immobilize Li^+ and lower its mobility, indicating the existence of an optimal PEO content. Tsao [70] synthesized and investigated the application of an ion-conductive and surface-active poly(ethylene glycol)-block-poly(acrylonitrile) (PEO-b-PAN) copolymer binder in LiFePO_4 (LFP) cathode materials. This binder not only improved the transport efficiency of lithium-ions in the cathode, but also significantly enhanced the discharge capacity and cycle stability

of batteries under high-rate conditions. For instance, the initial capacity reached 144 mAh g^{-1} at 0.5 C, with a capacity retention of approximately 98% after 200 cycles. Moreover, the increase in interfacial impedance was much lower than that of PVDF.

Dou et al. [71] synthesized a block copolymer comprising sodium alginate and Congo red. This copolymer established a continuous ionic channel via interconnected conductive bridges between the two block segments, thereby augmenting the ionic conductivity of the binder. David et al. [72] adopted the first and third design strategies, preparing a supramolecular ionic conductor with high ionic conductivity by using a polyether backbone and adding lithium salts.

Furthermore, we have also noticed that some researchers are using ion-conducting binders and expanding their application scale, such as the teams led by Meng et al. [73–75]. By using carbon dioxide-based polycarbonate polymer (PPC) and its derivatives, which can already be prepared on a large scale, as binders in solvent-free dry cathodes, this approach exhibits advantages such as environmental friendliness, low energy consumption, and high load capacity compared with the traditional wet process [76,77]. In addition, due to the abundant polar groups (e.g., carbonate groups, ether bonds, urethane bonds, etc.) in PPC-based polymers that coordinate with lithium-ions to provide ion transport channels, they can significantly improve the ionic conductivity of the electrode compared with the insulating PVDF. For example, the team [74] used a thermoplastic polymer poly(lactide-co-propylene carbonate) as a functional binder; the solvent-free fabricated LiFePO_4 cathode had a high areal loading of active material up to $57 \text{ mg}\cdot\text{cm}^{-2}$, and the battery assembled with this LiFePO_4 cathode and a lithium metal anode could deliver a specific capacity of $135.5 \text{ mAh}\cdot\text{g}^{-1}$ at 0.2 C. Similarly, Chen et al. [73] from the same team compared the performance of four polycarbonate-based binders (PPC, E-PPC, PPC-P, E-PPC-P) in dry LFP electrodes, and found that the chain-extended structure could significantly improve the tensile strength and ionic conductivity (up to $8.3 \times 10^{-5} \text{ S}\cdot\text{cm}^{-1}$). The prepared electrode had a loading capacity of $20 \text{ mg}\cdot\text{cm}^{-2}$, and the specific capacity of its half-cell was stably maintained at $140\text{--}150 \text{ mAh}\cdot\text{g}^{-1}$ at 0.5 C. Such PPC-based polymers with both high ionic conductivity and large-scale preparation potential are ideal binder systems that are compatible with the dry process and balance ion conduction, bonding strength, and green manufacturing.

2.2.2. Electron-Conducting Polymer Binder

Although current research on conductive adhesives has made progress, polymers have long been regarded as insulators because their covalent bond structure lacks free electrons. Due to the poor electronic conductivity of traditional binders [78,79], a certain amount of electronic conductors (e.g., conductive carbon black, carbon nanotubes) are usually added during the preparation of cathode sheets. To address this issue, scientists have developed electronic conductive binders. These binders directly construct electronic pathways through conductive material compounding or intrinsically conductive polymers, reducing the dosage of conductive agents and thus improving the energy density of batteries.

For a long time, polymers have generally been regarded as insulators, because covalent bonds in organic compounds are formed by sharing electrons from atoms to form local interatomic bonds, with no free electrons available for conduction. However, in 1977, Hideki Shirakawa et al. [80] discovered that iodine-doped polyacetylene exhibited metallic behavior and ultra-high electronic conductivity. To date, numerous electronic conductive polymers have been synthesized. In binder design, the core strategies for electronic conductive polymers include: constructing conjugated polymer backbones (e.g., PEDOT: PSS, polyaniline) to achieve delocalized electron conduction, and optimizing conductivity through chemical doping or self-doping (e.g., introducing sulfonic acid groups); combining cross-linked networks to enhance mechanical strength, while modifying with functional groups (e.g., thiol groups, PEO segments) to improve interfacial adhesion and electron transport; adopting composite strategies (e.g., compounding with carbon nanotubes, silver nanowires, or forming interpenetrating network structures) to construct three-dimensional conductive networks, balancing conductivity and flexibility.

For example, Zhang et al. [81] prepared a multifunctional polymer binder SAPPProDOT via the esterification reaction between the hydroxyl groups of sodium alginate (SA) and the carboxyl groups of 3,4-ethylenedioxythiophene-2,5-dicarboxylic acid (ProDOT). Owing to the conjugated network of PProDOT and lithium doping in the battery environment, SA-PProDOT became conductive and matched the conductivity required for LiFePO_4 LIBs without the need for conductive additives such as carbon black. Wang et al. [82] hybridized and cross-linked carbon nanotubes with cellulose nanosheets to obtain a composite binder with electron conductivity and robust mechanical strength for LCO cathodes. Without additional conductive additives, the electrode achieved an electronic conductivity of 816 mS cm^{-1} and supported ultrahigh-mass-loading electrodes with an LCO areal loading of up to 86 mg cm^{-2} and a corresponding areal capacity of 12.1 mAh cm^{-2} . At a loading of 30 mg cm^{-2} , the capacity retention reached 93% after 350 cycles at 0.5 mA cm^{-2} .

On the other hand, Seok et al. [83] prepared a highly conductive bifunctional binder PPP composed of PEDOT:PSS and PEG, for high-energy-density LFP cathodes. PSS was used to encapsulate PEDOT to improve its dispersibility, and PEG was further introduced to reduce the insulating coverage of PSS on PEDOT, thereby optimizing the dispersion of the binder and the electronic conductivity of PEDOT. As shown in Figure 6, the electronic conductivity of the optimized electrode reached 3.02 S cm^{-1} .

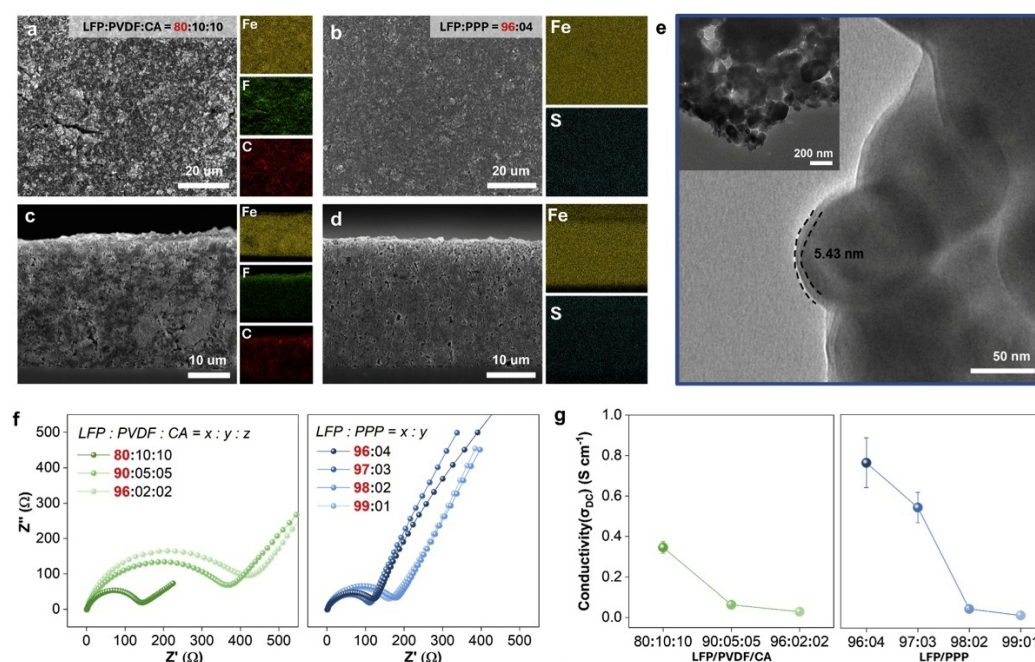


Figure 6. Comparative characterization of LFP cathodes prepared with PVDF and SuperC45, or with PEDOT:PSS/PEG (PPP) bifunctional binder. (a,b) Plane-view and (c,d) cross-sectional SEM images with corresponding EDS elemental mapping of (a,c) LFP:PVDF:SuperC45 (80:10:10), and (b,d) LFP:PPP (96:04). (e) HR-TEM images of LFP particles coated with a thin PPP bifunctional binder layer. (f) Nyquist plots from EIS results for LFP:PVDF:SuperC45 (80:10:10, 90:05:05, and 96:02:02) and LFP:PPP (96:04, 97:03, 98:02, and 99:01). (g) Conductivity plots of the same electrodes. [83]. (Reprinted with permission from Ref. [83]. Copyright 2026, Elsevier Inc.).

2.2.3. Ion-Electronic Double-Conductor Binder

To reduce the resistance of cathode components, a new class of organic polymers capable of conducting both ions and electrons, known as mixed ionic-electronic conductors (MIECs), has been proposed. These polymers significantly decrease the overall resistance of electrode composites while still providing the structural integrity of traditional binders.

For ionic-electronic dual-conductive binders, common molecular design strategies include the following: electrochemical doping of conjugated polymers to improve ionic conductivity—for example, the most extensively studied conductive binder is PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)) [84]; mixed ionic-electronic conductive block copolymers [85,86], where ionic conductive and electronic conductive phases are continuously connected, have also shown some practicality as binders for LFP cathodes; finally, introducing ionic conductive side chains onto electronically conductive conjugated backbones to facilitate lithium-ion transport is also a feasible strategy [87–89], whose effectiveness has been confirmed by numerous studies.

Meanwhile, alternative design approaches exist, as exemplified by the work of Pace et al. [90,91], who demonstrated a strategy for mixed conductive binders, where conjugated polymers are compounded with non-conjugated polyelectrolytes through electrostatic interactions to act as ionic cross-links. This endows the composite with mechanical strength, prevents its dissolution in common battery electrolytes, and exhibits high electronic and ionic conductivities. For example, Pace et al. [90] employed electrostatic cross-linking between an anionic conjugated polyelectrolyte and a cationic polymeric ionic liquid to prevent dissolution and detachment of the binder, forming a cohesive mixed-conductive binder. It exhibited an electronic conductivity of 1 S cm^{-1} and an ionic conductivity of $6 \times 10^{-8} \text{ S cm}^{-1}$. A capacity retention of 63% was achieved after 400 cycles at 0.5 C, whereas that of PVDF was only 6%.

2.3. Self-Healing Functional Binder

Binders play a crucial role in adhering active materials, conductive agents, and current collectors in electrodes. Traditional binders (e.g., PVDF) are insulating mechanical support materials; however, during long-term cycling, the volume expansion/contraction of electrode materials caused by lithium-ion intercalation/deintercalation leads to microcracks in the binder layer, damaging the structural integrity of the electrode. This triggers problems such as active material detachment and conductive network fracture, ultimately resulting in capacity fading and shortened cycle life. Self-healing binders can achieve autonomous crack repair through dynamic chemical bonds or supramolecular interactions, which helps maintain electrode structural stability and has become an important research direction for improving the durability of high-energy-density batteries.

The primary driving force for self-healing stems from intramolecular or intermolecular interactions, such as hydrogen bonding. Furthermore, as linear molecular architectures are incapable of providing sufficient binding strength, constructing three-dimensional binder networks through physically and chemically cross-linked structures—integrated with self-healing functionality—represents a promising strategy to accommodate materials undergoing substantial volume changes. Cross-linked frameworks provide the physicochemical foundation for self-healing: such architectures form a three-dimensional network wherein dynamic chemical bonds can reform after fracture, thereby enabling autonomous repair. For instance, Jin et al. [92] achieved *in-situ* cross-linking at room temperature through reconfigurable hydrogen bonds and ionic bonds by cross-linking polydopamine, phytic acid, and poly(acrylamide-co-2-(dimethylamino)ethyl acrylate), and the resulting binder exhibited excellent self-healing capabilities. Wei et al. [93] synthesized a self-healing supramolecular binder additive TD (polythioctic acid-co-diisopropenylbenzene) for Ni-rich layered oxide cathodes in high-voltage solid-state lithium batteries. TD forms a reversible cross-linked network via dynamic covalent disulfide bonds and carboxyl hydrogen bonds, which accommodates volume strain in the cathode and repairs cracks. Moreover, the disulfide bonds decompose under electrochemical oxidation, *in situ* generating a sulfur-rich, dense cathode electrolyte interphase (CEI). This interphase suppresses transition metal dissolution and LiTFSI decomposition, stabilizing the cathode/electrolyte interface. Under lean electrolyte conditions (E/C ratio = 1.8 g Ah⁻¹), a 3 Ah pouch cell delivers an energy density of 258 Wh kg⁻¹, with a capacity retention of 82.6% after 500 cycles at 1 C.

Liu et al. [94] synthesized a polymer binder FHCP cross-linked by both hydrogen bonds and disulfide bonds. By forming a cross-linked network, this polymer exhibits good self-healing capabilities. As can be seen from Figure 7, by comparing the stress-strain curves of the original and self-healed binders, it is confirmed that FHCP-10 can achieve a strain recovery rate of 97.8% and a stress recovery rate of 91.9% due to the synergistic effect of dynamic disulfide bonds and hydrogen bonds, demonstrating excellent self-healing efficiency and endowing it with great application potential in long-term battery operation.

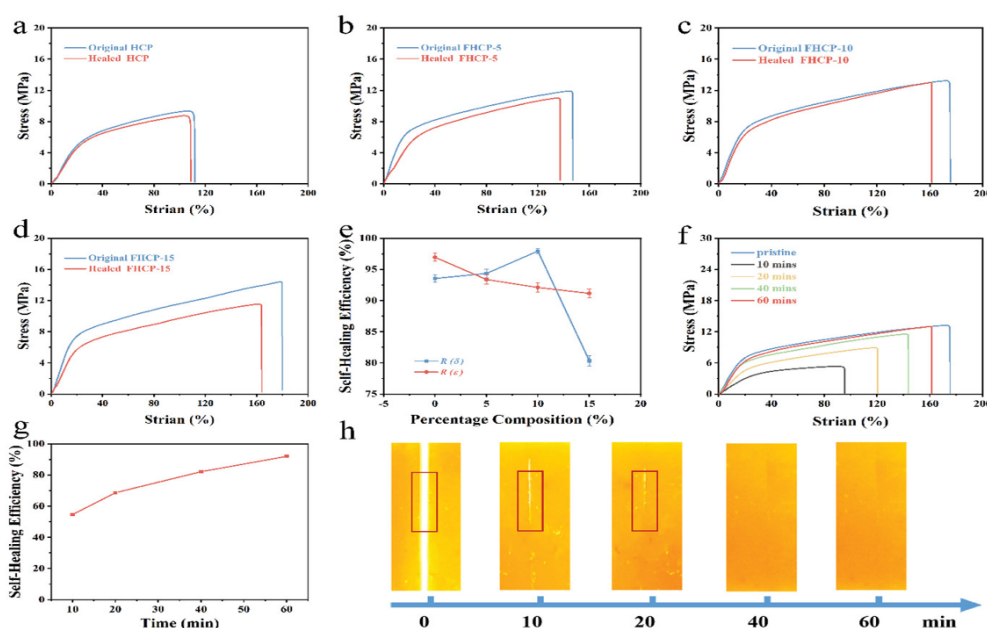


Figure 7. Stress–strain curves of original and healed (a) HCP; (b) FHCP-5; (c) FHCP-10; and (d) FHCP-15. (e) Effect of HCCP content on self-healing efficiency of FHCP. (f) Stress–strain curves and (g) self-healing efficiency diagram of FHCP-10 after different repair times. (h) Optical photos of FHCP-10 after different repair times [94]. (Reprinted with permission from Ref. [94]. Copyright 2022, Wiley-VCH.)

2.4. High Voltage Resistant Binder

With the advancement of portable electronic devices and related technologies, the application of high-voltage transition metal oxide cathodes (TMOCs) has been promoted, such as LiCoO_2 (LCO) [95]. However, this poses challenges in terms of high voltage. These problems include rapid capacity fading and low Coulombic efficiency, especially at high temperatures, resulting from severe oxidative decomposition of electrolytes, failure of active materials, and dissolution of transition metal ions from active materials [96–98]. The mitigation of these issues will mainly lie in how to effectively establish a multi-scale compatible cathode-electrolyte interface (CEI) when traditional carbonate electrolyte systems are still in use. To this end, the development of advanced polymer binders for high-voltage TMOCs should be one of the most effective strategies.

In the design of high-voltage-tolerant cathode binders, there are the following design strategies:

Firstly, improving electrochemical compatibility. In general, excellent electrochemical compatibility is essential for binders, which ensures their structural stability to achieve basic binding functions and enables excellent battery performance including high initial Coulombic efficiency. For example, Dou et al. [99] synthesized a hydrogen bond-based cross-linked aqueous binder using sodium lignosulfonate and guar gum as raw materials. Due to the abundant sulfonic acid groups in sodium lignosulfonate, it has good antioxidant capacity, thus achieving a charge-discharge voltage of 3–4.6 V, which is higher than the normal 3–4.2 V of lithium cobalt oxide. Binders for high-voltage TMOCs based on electrochemical compatibility can be divided into thermodynamically stable binders and kinetically stable binders. The former means that the binder provides high antioxidant capacity and thus will not lose electrons and be oxidized, represented by polyacrylonitrile (PAN) and polyvinyl butyral (PVB). The latter means that such binders are thermodynamically unstable themselves, but through unique chemical mechanisms on the surface of active particles, they can induce the formation of an effective CEI layer to obtain kinetic stability during repeated cycling, represented by styrene-butadiene rubber (SBR) [100].

Secondly, improving the dispersion capacity of electrode components and enabling uniform coverage on the surface of active materials. The uniform coverage of binders on the surface of TMOCs is conducive to kinetically inhibiting electrolyte oxidation and generating a thin and effective CEI layer, which can significantly improve cycle stability under high-voltage operation. Uniform dispersion facilitates electron and ion transport, thereby achieving high power output [101]. For example, chen et al. [102] adopted a thermal pulse sintering strategy to modify sodium carboxymethylcellulose (CMC), in which the polar groups such as carboxyl groups ($-\text{COOH}$) and hydroxyl groups ($-\text{OH}$) in CMC were controllably removed and converted into a continuous O-doped carbon network rich in ether bonds ($\text{C}-\text{O}-\text{C}$). This modification significantly improved the fluidity and film-forming property of the binder, enabling it to form a uniform and continuous coating layer with a thickness of approximately 4 nm on the surface of LiCoO_2 particles, thus allowing its application in 4.6 V LiCoO_2 cathodes, as shown in Figure 8. In terms of battery performance, it exhibited a capacity retention of 93% after 200 cycles at 1 C, while the unmodified CMC-based cathode only had an initial specific capacity of $185 \text{ mAh}\cdot\text{g}^{-1}$ with rapid capacity fading, showing a significant improvement by comparison.

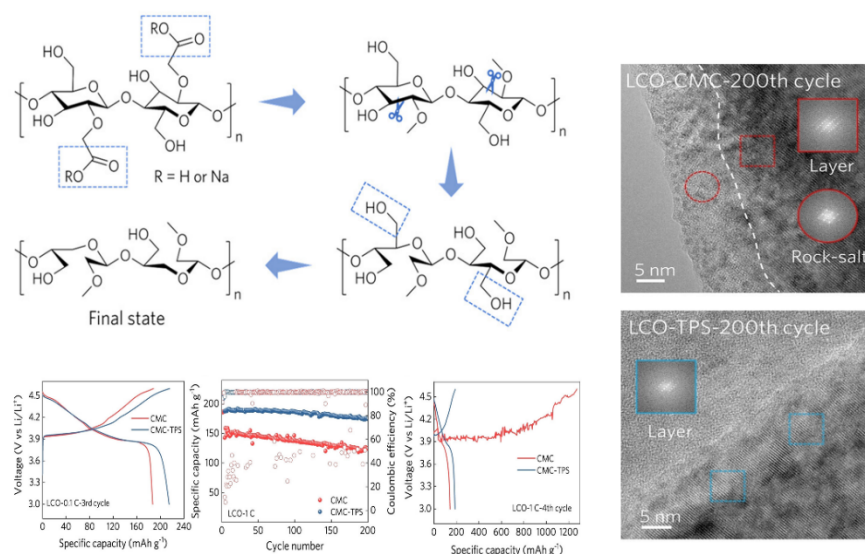


Figure 8. Schematic diagram of CMC-TPS polymer formation; electrochemical performance and HRTEM images of LCO cathodes with various binders after 200 cycles [102]. (Reprinted with permission from Ref. [102]. Copyright 2025, Wiley-VCH.).

Thirdly, enhancing the chelating capacity for transition metal ions. Under high-voltage operation, the crystal structure of cathode materials may collapse, leading to the dissolution of transition metal ions (e.g., Ni, Co, and Mn) into the electrolyte. These dissolved metal ions will exacerbate side reactions at the CEI and affect battery performance. Therefore, designing binders capable of forming stable chelates with transition metal ions is crucial for improving battery performance. For instance, Huang et al. [103] used lithium dextran sulfate (DSL) as a binder for LCO cathodes. The excellent interaction between the sulfate groups of DSL chains and LiCoO₂ particles enhanced the stability of Co-O chemical bonds, further inhibiting the harmful phase transition from O3 to H1-3 above 4.55 V. In contrast, Luo et al. [104] used cyanoethyl cellulose (CEC) as the binder for LCO cathodes. The highly polar cyano side chains not only improve the lithium-ion diffusion rate but also effectively chelate transition metal ions to suppress their dissolution. The CEC binder promotes the formation of a thin and dense LiF-dominated cathode electrolyte interphase (CEI) through cyano and hydroxyl groups, which inhibits carbonate solvent decomposition and reduces interfacial impedance. Meanwhile, the cyano groups chelate Co ions to suppress their dissolution, thereby significantly enhancing the cycling stability at a high voltage of 4.6 V. Specifically, it exhibited an initial specific capacity of 165.7 mAh·g⁻¹ and a capacity retention of 95.9% after 200 cycles at 4.6 V and 1 C, and its rate performance at 5 C was superior to that of PVDF.

3. Conclusions and Perspective

3.1. Future Outlook

Despite the significant progress made in the molecular design of polymer-based cathode binders, they still face numerous key scientific and engineering challenges for practical application and next-generation high-energy-density batteries. Future research should focus on the following specific and implementable development directions:

- (1) Combine machine learning, molecular dynamics simulations, and high-throughput experiments to establish the structure-activity relationship between functional group types, molecular chain structures, and bonding properties/electrochemical stability. This will enable efficient, accurate, and low-cost molecular design, accelerating the development cycle of new functional binders.
- (2) In-depth study the interaction mechanisms between binders, electrolytes, and the CEI interface. Develop multifunctional binder systems that can simultaneously stabilize the cathode interface, regulate the solvation structure, and inhibit side reactions, achieving integrated optimization of the cathode-electrolyte interface.
- (3) Aiming at the volume effect, transition metal dissolution, and interface instability of high-nickel/lithium-rich manganese-based cathodes, design binders with strong anchoring, oxidation resistance, and self-healing properties. Meanwhile, develop specialized binders for solid-state batteries compatible with sulfide and oxide solid electrolytes to improve solid-solid interface contact and enhance interfacial ion transport and structural stability.
- (4) Facing practical application needs, focus on developing binders that still possess high bonding strength, excellent ion transport, and structural stability under extreme conditions such as high areal loading, lean electrolyte, and fast charging. Solve problems such as electrode cracking, blocked ion transport, and cycle life decay.
- (5) Develop water-soluble, fluorine-free, and environmentally friendly binders to replace traditional PVDF systems. Simplify synthetic routes, reduce raw material costs, and develop scalable slurry preparation and film-forming processes, promoting the transformation of binders from laboratory research to industrial application.

3.2. Conclusions

This review summarizes the latest progress in the molecular structure design of polymer-based cathode binders, emphasizing their transformation from “passive bonding” to “active functional regulation”. By introducing polar functional groups, conductive units, and dynamic bonding strategies, the conductivity, mechanical stability, and interface compatibility of electrodes have been significantly improved. Self-healing binders repair microcracks through dynamic networks to extend battery life, while high-voltage-tolerant binders inhibit electrolyte decomposition by optimizing interface chemistry to enhance the cycling stability of high-voltage cathodes. A summary of the content is shown in Table 1.

In the future, function-oriented polymer binders will become key materials for high-energy-density, long-cycle, and high-safety lithium-ion batteries and all-solid-state batteries. Through interdisciplinary innovation and engineering breakthroughs, new cathode binders are expected to provide core support for the next generation of high-performance energy storage systems.

Table 1. Summary table of functional classification and key performances of polymer-based cathode binders.

Functional Classification	Specific Classification	Binder System	Active Material	Key Performance
Improving Bonding Strength	Forming Hydrogen Bonds	Hydroxyl-rich siloxane nanohybrid (SNH) [25]	NCM811	The cohesion of SNH is 202 gf/12 mm, which is significantly higher than that of PVDF slurry (120 gf/12 mm).
		H-P composite binder [27]	LFP	The peel strength of the H-P electrode is 23.23 N·m ⁻¹ , which is much higher than that of PVDF (0.322 N·m ⁻¹); the capacity retention rate is 86.3% after 500 cycles at 1 C.
	Electrostatic Interaction	PVDF-CTFE [39]	NCM712	For the assembled pouch battery, the initial capacity is 198.5 mAh·g ⁻¹ at 0.1 C; the capacity retention rate is 74.5% after 300 long cycles at 0.5 C.
	Controlling crystallinity	PVDF [50]	LiNi _{0.88} Co _{0.06} Mn _{0.06} O ₂	The capacity retention rate is 83.7% after 100 cycles at 0.5 C.
	Formation of cross-linked structures	PNAL [53]	NCM811	A capacity retention rate of 75.7% is achieved after 300 cycles at 3 C and 4.6 V.
	Formation of coordination bonds	PI [61]	NCM811	The peel strength is 0.571 N·mm ⁻¹ ; the capacity retention rate is 78.6% after 100 cycles at 4.7 V and 0.2 C, and the discharge capacity is 164.4 mAh·g ⁻¹ at 5 C.
Enhancing Conductivity	Ion-conducting polymer binder	PI [65]	LFP	The lithium-ion diffusion coefficient is 7.23 × 10 ⁻¹⁰ m ² /s, and it can cycle stably for 200 times at 0.5 C.
		PTFE [66]	NCM712	The ionic conductivity of the polymer is 1.6 × 10 ⁻⁵ S·cm ⁻¹ , and the ionic conductivity of the composite cathode is 4.1 × 10 ⁻³ S·cm ⁻¹ ; the discharge specific capacity reaches 180.7 mAh·g ⁻¹ at 0.1C, and the capacity retention rate is 90% after 300 cycles at 0.5 C.
	Electron-conducting polymer binder	PEDOT:PSS/PEG [83]	LFP	The electronic conductivity reaches 3.02 S/cm; the capacity of the half-cell is 131 mAh·g ⁻¹ at 8 C, and the capacity retention rate remains 96% after 1000 cycles at 3 C.
	Ion-Electron Dual Conductor	CPC [90]	LFP	The electronic conductivity is 1 S/cm, the ionic conductivity of the polymer is 6 × 10 ⁻⁸ S·cm ⁻¹ , and the capacity retention rate is 63% after 400 cycles at 0.5 C.
Self-Healing Binder	Forming Cross-Linked Structure	Polyipoic acid-co-diisopropenyl benzene [93]	NCM811	The energy density of the 3 Ah pouch battery reaches 258 Wh·kg ⁻¹ , and the capacity retention rate is 82.6% after 500 cycles at 1 C.
High-Voltage Resistant Binder	Improving Dispersion Capacity	CMC [102]	LCO	The capacity retention rate is 93% after 200 cycles at 4.6 V and 1 C.
	Enhancing Chelation	Cyanoethyl cellulose (CEC) [104]	LCO	The capacity retention rate is 95.9% after 200 cycles at 4.6 V and 1 C (initial capacity: 165.7 mAh·g ⁻¹).

Author Contributions

K.Z.: writing—review & editing, writing—original draft, visualization, validation, methodology, investigation, funding acquisition, formal analysis, conceptualization; Z.Z.: writing—original draft, methodology, investigation, formal analysis; L.Z.: writing review & editing, visualization, validation, supervision, project administration, conceptualization; D.H.: writing—review & editing, visualization, validation, supervision, funding acquisition, conceptualization. All authors have read and agreed to the published version of the manuscript.

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Informed Consent Statement

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Data Availability Statement

No new data were created or analyzed in this study.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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